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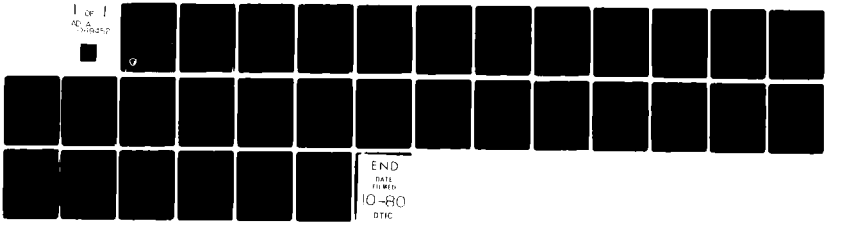

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'IN SITU' MEASUREMENT OF THE RATIO OF AEROSOL ABSORPTION TO EXTINCTION COEFFICIENT

AUGUST 1980

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20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*

A knowledge of the proportion of absorbed to scattered radiation for countermeasure smokes is necessary for modeling battlefield effects. Measurement of the total scattered component, however, is difficult. This report presents results obtained by using a new and simpler approach focussing on the extinction and the absorption coefficients (the total scattering coefficient is the difference quantity). Measurements of the absorption and extinction coefficients due to smoke produced by the combustion of red

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20. ABSTRACT (cont)

phosphorous[4] are simultaneously obtained in a small test chamber. The same CO_2 laser beam at λ = 9.55μm is used to produce both results. A form of aerosol spectrophone cell was used to measure the absorption coefficient while functioning as a transmission cell from which the extinction coefficient was calculated. The ratio of the absorption to extinction coefficients was then compared with estimates of the same quantities based on calculated particle sizes, the particle size estimates being obtained from characteristics of the settling process. The ratio agreed with that predicted within the experimental error.

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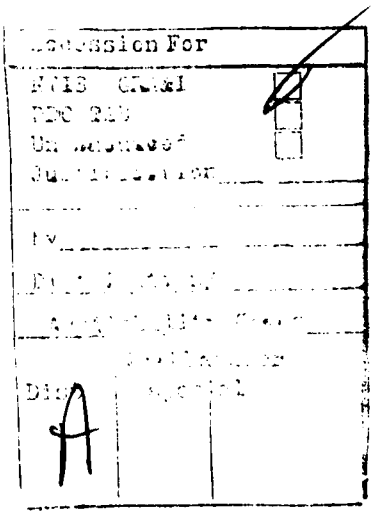

INTRODUCTION

Modeling of infrared (IR) propagation properties through atmospheric aerosols is of continuing interest for radiative transfer, atmospheric sensing, and communication purposes. The extinction coefficient is often used to describe the linear radiation losses[1] though an adequate description may require a knowledge of component quantities, the absorption and the scattering coefficients (refractive effects being treated separately[2,3]).

Absorption of radiation by both atmospheric gases and/or particles may be measured in situ by using spectrophones developed and used by the authors for a variety of environmental chamber and field measurements.[4-8]

Measurement of the total scattered component, though, is difficult since all angles must be included (or their omission justified) and detected with equal sensitivity.

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[1]J. E. A. Selby and L. A. McClatchey, 1976, Atmospheric Transmittance from 0.25 to 28.5 m: Computer Code LOWTRAN 3b, AFGL-TR-76-0258, Air Force Geophysics Laboratory, Hanscom AFB, MA

[2]S. Q. Duntley, 1948, "The Reduction of Apparent Contrast by the Atmosphere," JOSA, 38

[3]W. E. K. Middleton, 1952, Vision Through the Atmosphere, University of Toronto Press

[4]C. W. Bruce, 1976, Development of Spectrophones for CW and Pulsed Radiation Sources, US Army Electronics Command, Report ECOM-5802

[5]C. W. Bruce and R. G. Pinnick, 1977, "In-Situ Measurements of Aerosol Absorption with a Resonant CW Laser Spectrophone," J Appl Opt, 16:1972

[6]C. W. Bruce, R. J. Brewer, Y. P. Yee, and D. Bruce, 1978, "In-Situ Measurements of Aerosol Absorption," Proceedings 3rd Conference on Atmospheric Radiation," American Meteorological Society, Boston, MA

[7]C. W. Bruce, Y. P. Yee, B. D. Hinds, R. J. Brewer, and J. Minjarez, "In-Situ Measurements of Atmospheric Absorption," submitted J Appl Meteorol

[8]C. Samuel, C. Bruce, and R. Brewer, 1978, Spectrophone Analysis of Gas Samples Obtained at Field Site, US Army Electronics Research and Development Command, Report ASL-TR-0009

An alternative approach[3] is to simultaneously obtain the extinction coefficient by using a direct transmission measurement and the absorption coefficient by using an in situ spectrophone, the scattering coefficient then being the difference. In this method, the same optical path and beam power profile are used for both measurements to avoid questions of optical system differences. This approach to obtaining the components of the extinction has been applied in environmental chamber measurements of 10μm propagation through the smoke which is produced when red phosphorous is ignited. The measurements include correlations with estimates of the particle size and number densities and comparison with calculations of the propagation properties based on the data.

EXPERIMENTAL PROCEDURE

A spectrophone absorption cell was constructed for the in situ measurements discussed in this report. In the cell, smoke was generated and circulated to ensure a well-mixed medium. Absorption and extinction coefficients were then measured as a function of time for an extended period. The resulting settling process provides a continuously changing size distribution evolving toward smaller mean sizes and widths as well as lower total number densities for each size.

The measurements of this report were made at a CO_2 laser wavelength of 9.55μm.

Designs of the in situ gas/particle spectrophone system used have been described;[4,5] however, the nature of these measurements suggested a number of changes to the basic cylindrical system previously used. The measurements were expected to produce relatively high levels of absorption and extinction in a system in which mixing could take place.

Because of the corrosive nature of the substance, i.e., phosphoric acid, gold was used to replace aluminum or brass on exposed portions of the integral capacitance microphone. A stainless steel shield was installed to protect an optical window (BaF_2 wedge) at the end of the spectrophone nearest the smoke pellet. The cell could be removed for cleaning and purging before calibrating it with a gaseous absorber. Finally, a closed circuit circulation system was installed.

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[3]D. M. Roessler and F. R. Faxvog, 1979, "Optoacoustic Measurement of Optical Absorption in Acetylene Smoke," J Opt Soc Am, 12:1699

[4]C. W. Bruce, 1976, Development of Spectrophones for CW and Pulsed Radiation Sources, US Army Electronics Command, Report ECOM-5802

[5]C. W. Bruce and R. G. Pinnick, 1977, "In-Situ Measurements of Aerosol Absorption with a Resonant CW Laser Spectrophone," J Appl Opt, 16:1972

Figure 1 is a schematic diagram of the cell and optical system described. Details of the in situ spectrophone systems and signal processing for both pulsed and CW laser sources have previously been described.[4]

The procedure for settling measurements was to obtain a reference (presmoke) level of stabilized power on both of the calorimeters indicated in figure 1. The laser power was referenced to the input calorimeter for the duration of the measurements. The circulation pump was in operation during the pellet burning time and for about 2 minutes after this period. Subsequent mixing was due to the thermal gradients created by the absorption along the cylindrical axis. Absorption coefficient and cell input and output power were then recorded as a function of settling time. At very long settling times, the power approaches a final value representative of cell window losses and the absorption approaches zero. The cell output power may be slightly lower after the test because of particulate residue deposited on the windows. The cell contents are pumped out and air is inserted. The resulting output power and absorption values are then used as checks on the asymptotic values for the settling process. The agreement between the two results was good. The increased sustaining loss in transmission was due to particulate residue on the cell end windows. The residue accrued during or near burning time. In general, this loss was negligible whenever the window shielding cylinder was inserted.

The cell was calibrated by using ethylene gas, a substance whose absorption properties at 10μm laser wavelengths have been carefully measured by the authors. Spectrophones have high inherent sensitivity and, in normal use, the transmission loss across the cell is negligible. Percentage transmission losses in several other currently used ASL spectrophone systems, for example, range from about 10^{-5} to a maximum of unity (the latter for low visibility dust conditions). However, since larger fractional losses are required for accurate extinction measurements, the gaseous ethylene calibrations were performed over the range of the absorption coefficients measured during the settling process. During the calibration procedure, in which the acoustics were assumed not to be significantly altered by the smoke particles, the extinction and the absorption have essentially the same values since the molecular scattering is negligible. These corresponding values also provide a check on the accuracy of the extinction measurement. Figure 2 shows a sample calibration plot. The linearity is typically high when the power is referenced to cell (and microphone) center.

Again, the probe laser beam which is absorbed producing the acoustical signal is the same beam which is attenuated producing the measured extinction. The two measurements are therefore totally related quantities; i.e., they have identical beam power profile, location, and wavelength.

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[4]C. W. Bruce, 1976, Development of Spectrophones for CW and Pulsed Radiation Sources, US Army Electronics Command, Report ECOM-5802

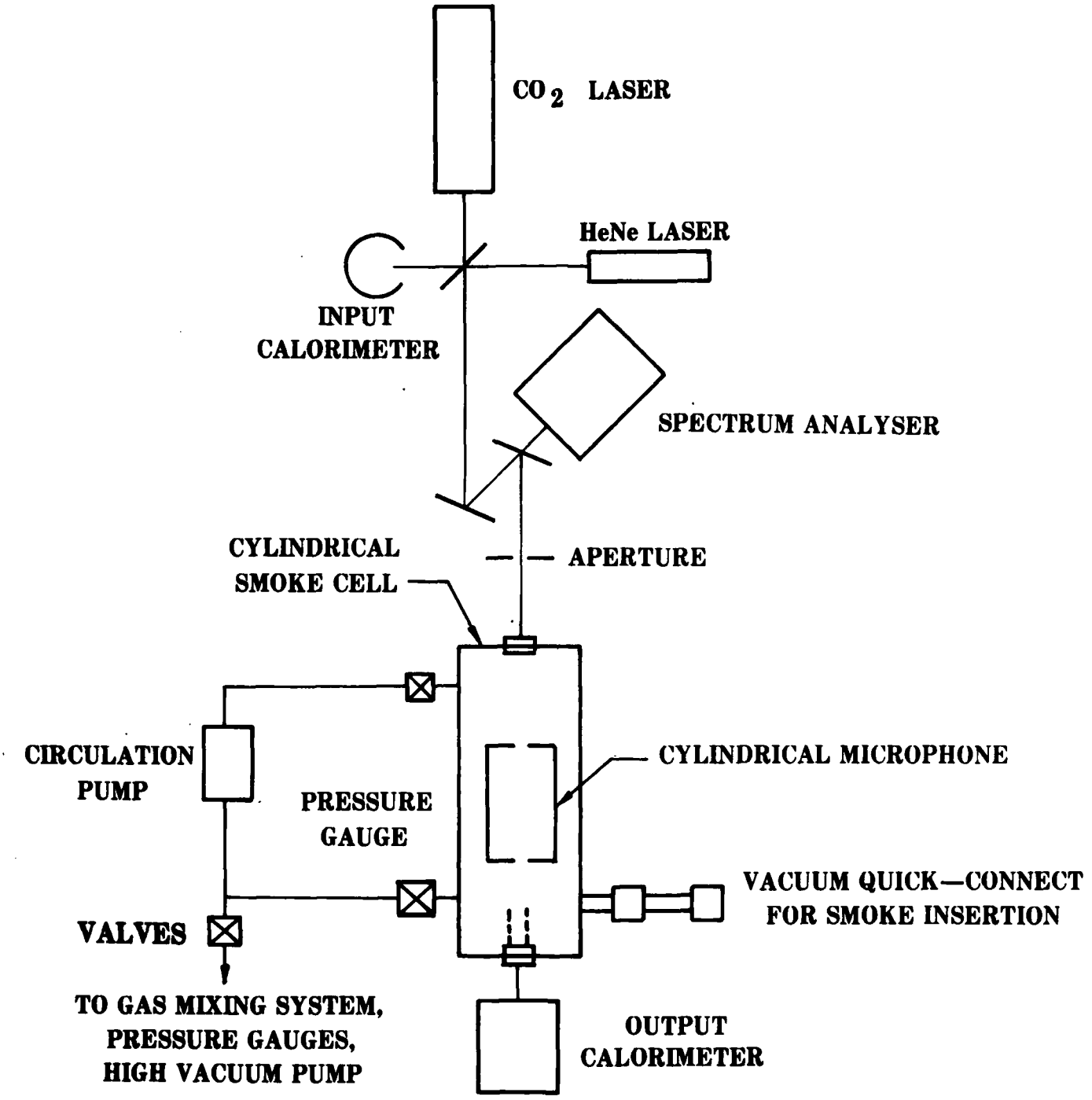

Figure 1. Optical system and particulate cell configuration.

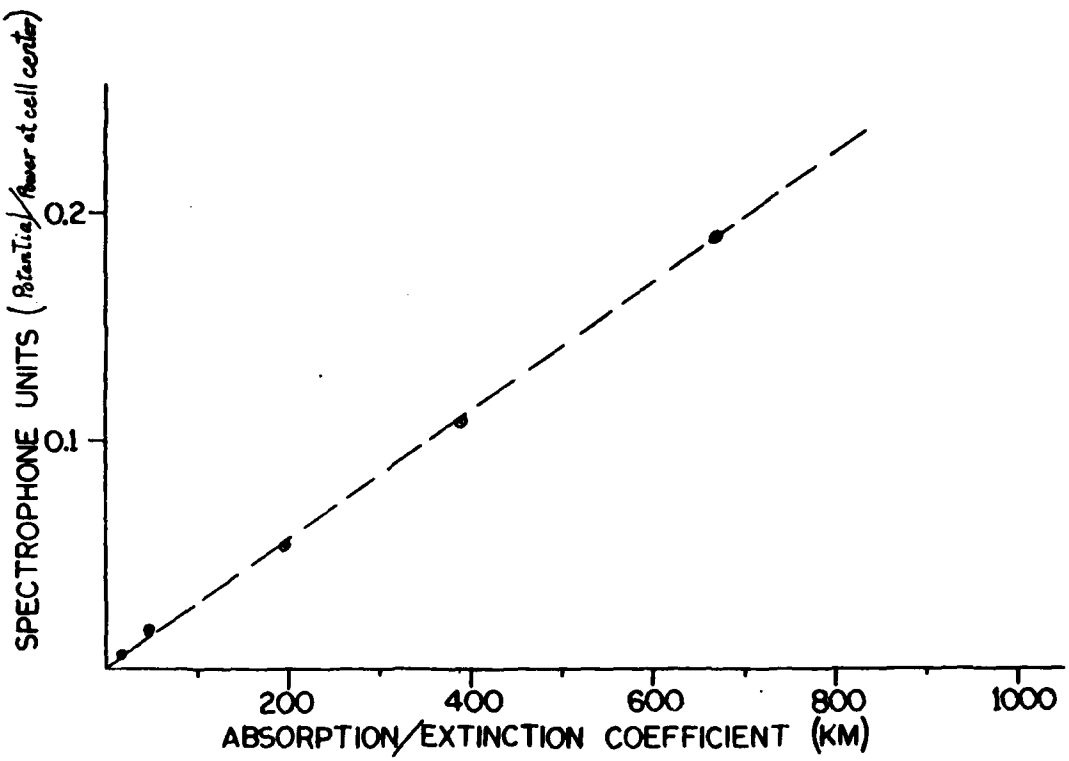

Figure 2. Sample ethylene calibration data in terms of cell output voltage divided by the total optical beam power at the center of the microphone (and at the position of the peak amplitude for the acoustical mode used) as a function of the measured extinction.

RESULTS AND ANALYSIS

A sample settling process for the smoke used is shown in the exponential representation of figure 3. The scattered proportion decreases slightly as a function of decreasing particle size (as the Lorenz-Mie theory predicts) with increasing settling time.

Positive curvature in both curves suggests a polydispersion of the smoke particles, while the (slight but real) reduction of this curvature at later times indicates a narrowing of the distribution with time, which is also expected from theory.

Since the signal of the cell output calorimeter may include a significant amount of radiation that has been scattered by the smoke particles, the forward scattering correction[10] was calculated for the wavelength and range of sizes encountered (estimates of these sizes will be discussed). The calculations show that the correction is negligible and that there is considerable latitude with respect to size in this conclusion. For the beam geometry of these measurements, the limiting radius for which corrections may be ignored is about 10μm.

To estimate the radii of particles most representative of the absorption/extinction as a function of time, calculations were performed from parameters of the settling process. Calculations rather than measurements were used since available particle counters were not felt to be suitable.

An estimate of the radius contributing the largest incremental absorption value ($\tilde{r}$) may be obtained from the settling curve slope. A spread or breadth of significantly contributing sizes can also be inferred, e.g., from the second derivative. Since this spread is simply an estimate, we will assume the smoke aerosol to be monodisperse.

First, we will assume that Stokes' law is an adequate approximation so that the terminal velocities, v_t, for particles moving under the force of gravity are given by

$$v_t = (2/9)\ \rho_m r^2 g/\eta\ , \qquad (1)$$

where ρ_m is the density of the particulate material, r is the particle radius, g is the acceleration due to gravity, and η is the viscosity of the medium. The assumption of a viscous medium is not precise since particle radii of less than 1μm are present. Then, two assumptions are made about the settling process in the chamber. These assumptions may seem contradictory, but actually they are compatible: (1) particles are lost to the bottom of the

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[10] A. Deepak and M. A. Box, 1978, "Forwardscattering Corrections for Optical Measurements in Optical Media. 1: Monodispersions," J Appl Opt, 17:2900

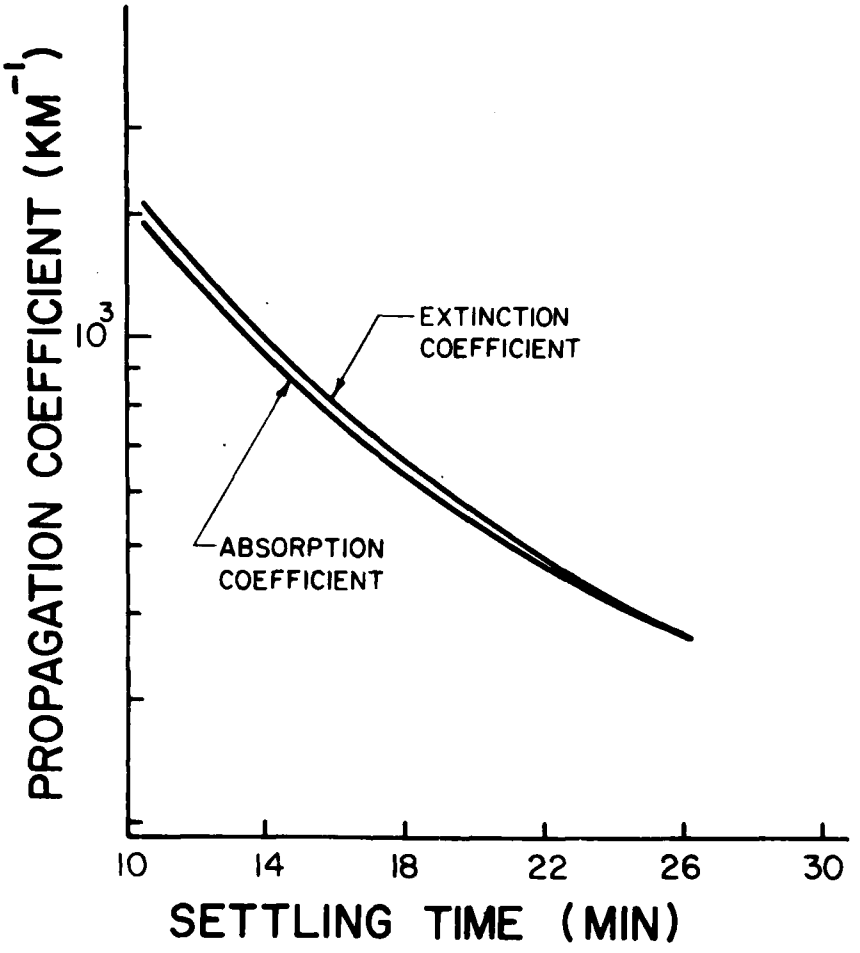

Figure 3. Extinction and absorption coefficients as a function of settling time in the cell of figure 1.

chamber at rates representative of those in still air, and (2) the particle number density distribution is independent of the vertical position in the chamber. The latter assumption is associated with the effect of the laser beam on the medium, i.e., that of a mild but continuous convection current. In addition we will treat the medium as monodisperse, though the treatment can be broadened by assuming a particle size distribution and integrating.

The rate of change of number density is then given by

$$\frac{dn}{dt} = -n\, Av_t \frac{1}{V} , \tag{2}$$

where n is the particle number density and A and V are the cross-sectional area and volume of the chamber.

Then integrating equation (2) gives

$$n = n_o e^{-\left[\frac{A}{V} v_t (t-t_o)\right]} , \tag{3}$$

or, if written to display the fact that this relation is for a given radius,

$$n(a) = n_o(a) e^{-\left[(2/9 \frac{A}{V} \frac{\rho_m g r^2}{\eta} (t-t_o)\right]} . \tag{4}$$

A/V represents an effective inverse height of the chamber. The second assumption means that the settling process takes infinite time, whereas without it the settling time is the effective height divided by the terminal velocity.

We assume that the number density, n, is proportional to the absorption coefficient, σ_a. The validity of this assumption is largely dependent on the criterion $2\pi r << \lambda$, but is less stringent than it would be for the extinction coefficient.[11] The calculated radii at the times of most interest (longest possible in the measurement) do adequately satisfy this criterion according to Lorenz-Mie dependencies.

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[11]S. G. Jennings, R. G. Pinnick, and H. J. Auvermann, 1978, "Effects of Particulate Complex Refractive Index and Particle Size Distribution Variations on Atmospheric Extinction and Absorption for Visible Through Middle IR Wavelengths," J Appl Opt, 17:1922

Then the estimate for the radius with the largest contribution to the absorption coefficient, σ_a, is

$$\tilde{r} = \left[\frac{9}{2} \frac{V}{A} \frac{\eta}{\rho_m g} \frac{\ln \sigma_a / \sigma_{a_o}}{t - t_o} \right]^{1/2} ,$$

or

$$\tilde{r} = (C \cdot S)^{1/2}, \tag{5}$$

where S = the slope of the settling curve and

$$C = \frac{9}{2} \frac{V}{A} \frac{\eta}{\rho_m g} . \tag{6}$$

Note that the radius values obtained from the relation derived without the second assumption (b, mixing) are given by

$$r = C^{+1/2} t^{-1/2} , \tag{7}$$

where t is the total settling time.

Here, there is no interaction with the data taken. The singularity at t = 0 is not of concern since all of the times of interest are relatively long. The results of calculations of radii using equation (7) prove to be of the same order of magnitude but, as expected, the radii are larger than those obtained for the model which incorporates mixing. Equation (7) will be used to represent an estimate of the maximum radius, r_{max}, present in the chamber as a function of time.

This analysis could be carried one step further to obtain estimates of the width of the absorption curve as a function of radius by integrating the relation

$$\sigma_a(r) = \sigma_{ao}(r) e^{-cr^2(t-t_o)}$$

over radii and assuming a form for the distribution $\sigma_a(r)$. The distribution will not be known a priori, but calculations using Gaussian and power law dependencies have been used in this study. Breadths of the order of 0.1μm number density at half width were obtained with changes over the measurement

period (at the conclusion of circulation to about 60 min later) that were of an order of magnitude smaller. Size distribution measurements were also attempted for similar settling runs by using a commercial PMS^{R} Model CSASP ($0.22\mu m < r < 15\mu m$) particle counter. These measurements were not considered conclusive since the size range was inadequate for these purposes.

By using the Lorenz-Mie theory and the complex index[12] for the chemical content of the smoke (phosphoric acid assumed but currently not verified), including growth of the particles as a function of relative humidity,[13] we computed the absorption-to-extinction ratios as a function of particle radius for various settling times. For these calculated results, the particles were assumed to be spherical and to consist of homogeneous mixtures of water and phosphoric acid.

Diffusion of the smoke particles to the wall was computed separately as a correction to the settling process, but the fractional loss rate was only 0.04 for 0.01μm radius particles and progressively smaller for larger radii.

Table 1 is an abbreviated version of the results of figure 3 as well as the absorption/extinction coefficient ratio as a function of settling time. These ratios are compared through the estimated particle radii to the Lorenz-Mie results. Calculated results were based on settling theory with mixing, settling theory without mixing for an estimate of particle maximum radii, and independent estimates by others[14] whose value depends only on the particle maximum radius. The estimated initial number density might be adequately high to produce a significant amount of multiple scattering, but no quantitative results were attempted with the minimal information on densities. Further, there is no indication of the effects of multiple scattering in the results.

A rough calculation of the absorption coefficient using mass loss in sample pellets during the burning process, the change in particle complex index due to growth with relative humidity, and an approximation of the Lorenz-Mie theory based on the relatively small size parameters ($2\pi r/\lambda << 1$) resulted in a value of absorption coefficient within a factor of two of the measured value.

The smoke particles generated for these measurements were predominately small ($r_{max} < 1\mu m$) and possibly delicate with respect to sampling processes. Since counting and sizing of such particles were difficult, the parameters necessary for the comparison of measured and calculated ratios were derived from the propagation properties during the settling process. These measurements are

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[12]M. R. Querry and I. L. Tyler, 1978, "Complex Refractive Indices in the Infrared for H_3PO_4 in Water," J Opt Soc Am, 68:1404

[13]G. Hänel and K. Bullrich, 1978, "Physico-Chemical Property Models of Tropospheric Aerosol Particles," Beitrage Zur Physik der Atmosphäre, 51:129

[14]R. G. Pinnick and S. G. Jennings, private communication

TABLE 1. MEASURED AND CALCULATED RATIOS, R, OF ABSORPTION TO EXTINCTION COEFFICIENTS AT A WAVELENGTH OF 9.55μm AS A FUNCTION OF SETTLING TIME AND ESTIMATED RADII

Time (min)	$\tilde{r}$ (μm)	r_{max} (μm)	R_{meas}	R_{calc} ($\tilde{r}$)	$R_{calc'}$ (ref 14)
13	0.63	0.66	0.97	0.96	
18	0.53	0.56	0.99	0.97	0.915
23	0.49	0.49	0.99	0.98	

Legend:

r_{max} Calculated estimate of maximum radius

$\tilde{r}$ Calculated estimate of radius of maximum absorption

R_{meas} Measured ratio of absorption to extinction coefficients

R_{calc} Calculated values using above radii and the Lorenz-Mie theory

$R_{calc'}$ Ref 14 uses linearized simplification of Lorenz-Mie theory

expected to provide a simpler step en route to use of this technique for particulates having distributions with propagation effects predominately from larger radii. A cell with relatively long path, internal mixing system, relative humidity, smoke source mass loss measurements is being prepared for measurements of σ_a/σ_e ratios (σ_e = extinction coefficient) for atmospheric aerosol dusts. Particle counting and sizing measurements which might be appropriate and accurate for this application as also being investigated.

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